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# Development of Long-Life Lithium/Sulfur-Containing Polyacrylonitrile Cells

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Project ID # bat454

This presentation does not contain any proprietary, confidential, or otherwise restricted information

# Overview

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## Timeline

- Project start date: Oct. 2016
- Project end date: Sept. 2021
- Percent complete: 70%

## Budget

- Total project funding
  - DOE share \$ 50 M
  - Contractor share
- Funding for FY 2019: \$ 10 M
- Funding for FY 2020 (if available): \$ 10 M

## Barriers

- Barriers addressed
  - 500 Wh/kg Li-S battery
    - High loading sulfur cathode
  - Cycle life
    - Stable, high efficiency lithium anode
    - Solid electrolyte for lithium protection

## Partners

- Project Lead
  - PNNL
- National Laboratories
  - PNNL, INL, Brookhaven, SLAC/Stanford
- Academia
  - Binghamton, U. Washington, U. Texas

# Relevance

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- **Overall Battery 500 Objective**
  - Develop commercially viable Li battery technologies with a cell level specific energy of 500 Wh/kg through innovative electrode and cell designs that enable utilization of maximum capacity of advanced electrode materials
- **Chemistry**
  - Utilize a **Li metal anode** combined with a compatible electrolyte system, and either
    - A nickel-rich NMC or S
- **Keystone project (2): Electrode Architecture**
  - Leverages materials and chemistry advances from Keystone (1): Materials and Interfaces to enable stable, high loading anode and cathode
  - Provides component and materials support for Keystone (3) Cell Design and Integration

# Milestones: Keystone 2 and UCSD

End date	12/31/2019	03/31/2020	06/30/2020	09/30/2010
Keystone Project 2 Electrode Architecture	Report Li/SPAN coin cell results that can support a 250 Wh/kg, 2 Ah pouch design with 100 cycles.  <b>Completed</b>	Develop new 3D anode structures and test such using coin cell standard protocols to achieve 300-350 Wh/kg (cell-level) for 200 cycles.  <b>Completed</b>	Develop new polymer protective layers for Li anode, test and report such using coin cell standard protocols.  <b>On Track</b>	Demonstrate the performance of a SPAN based Li-S pouch cell for 300-350 Wh/kg.  <b>On Track</b>
UCSD	Report Li/SPAN coin cell results that can support a 250 Wh/kg, 2 Ah pouch design with 100 cycles.  <b>Completed</b>	Quantify the 3D morphology of cycled lithium anode under various pressure.  <b>Completed</b>	Identify a pathway to more than 350 Wh/kg (scaled to 2 Ah) using SPAN. Report SEI characterization of lithium anode with surface coatings.  <b>On Track</b>	Demonstrate the performance of a SPAN based Li-S pouch cell for 300-350 Wh/kg.  <b>On Track</b>

# Keystone 2 Challenges and Approaches

## 3D Lithium

3D hosts with stable interface with electrolytes



Modeling

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Analysis

Solid Electrolytes



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3D architecture



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**A combined experiment, modeling, and characterization effort to:**

- Understand how lithium is lost
- Model effect of pressure and other experimental factors
- Develop new solid electrolytes
- Design new 3D architectures to achieve high efficiency and long cycle life

**Thick cathode architecture  
> 6 mAh/cm<sup>2</sup> SPAN**

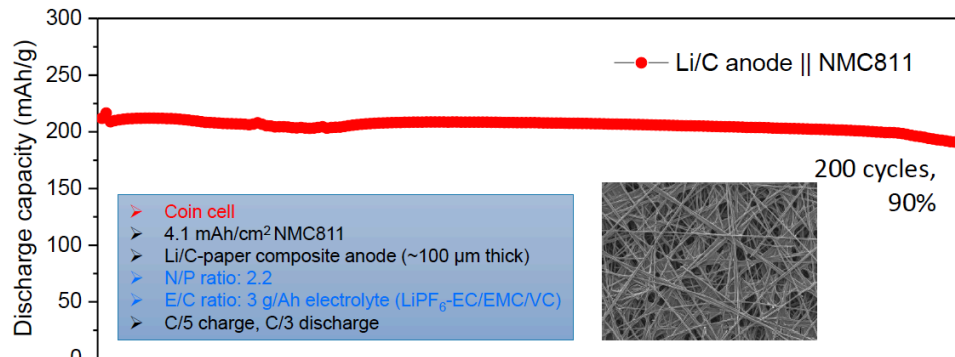
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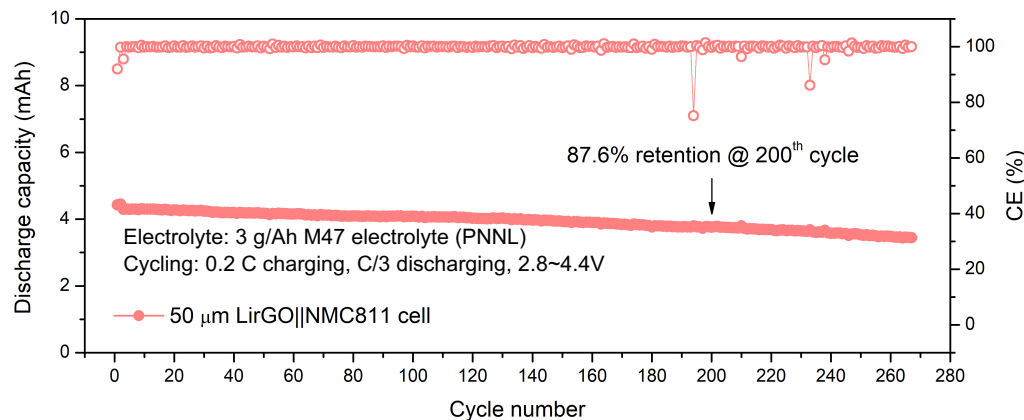
**Engineer high-loading SPAN electrode to maximize cell energy density**

- Develop electrolyte formulations that enable both Li and SPAN
- Optimize binder, porosity, and electrode conductivity
- Demonstrate lean electrolyte operation

# Technical Accomplishments: New 3D Li Architecture



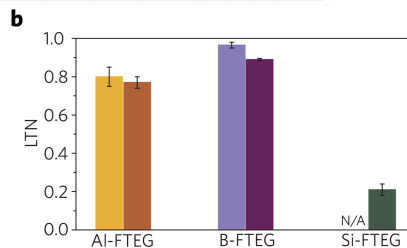
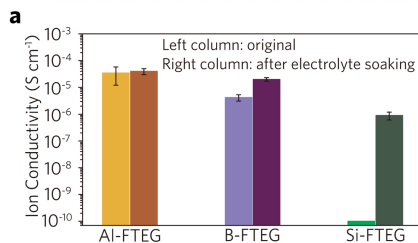
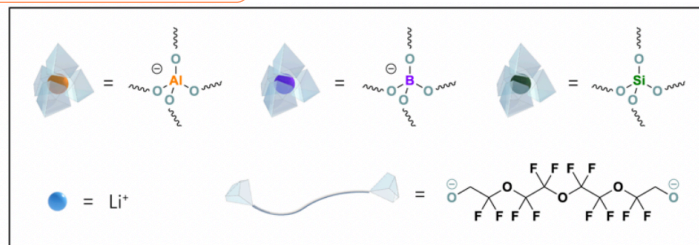
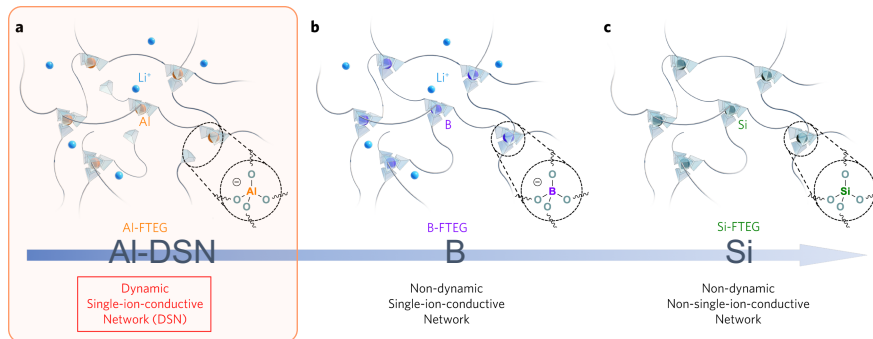
C. Niu



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H. Chen and H. Wang

# Technical Accomplishments : New Polymer Coating Layer



**Artificial SEI design with multifunction in a single matrix**

1. Dynamic flowability
2. Fast  $\text{Li}^+$  single-ion conduction
3. Electrolyte blocking

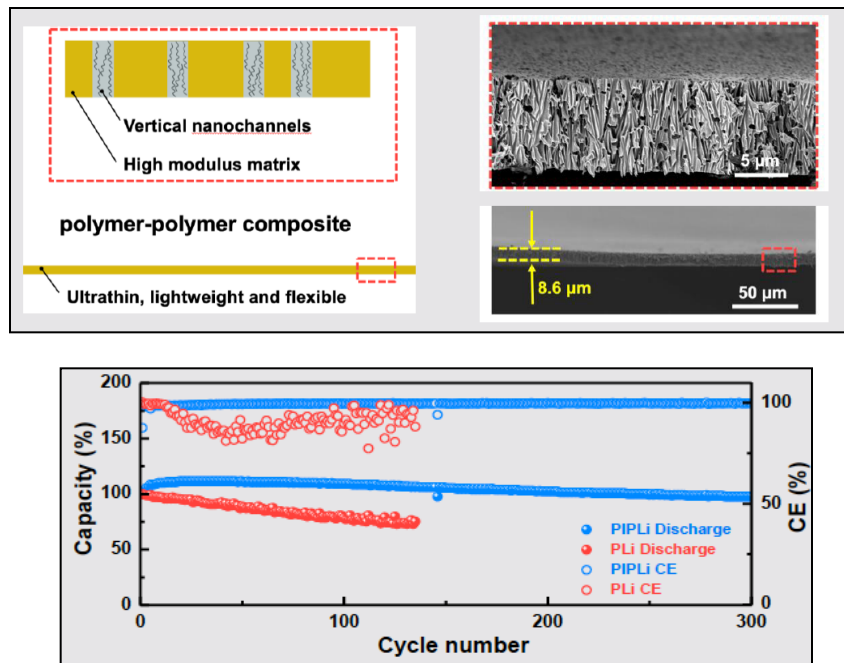
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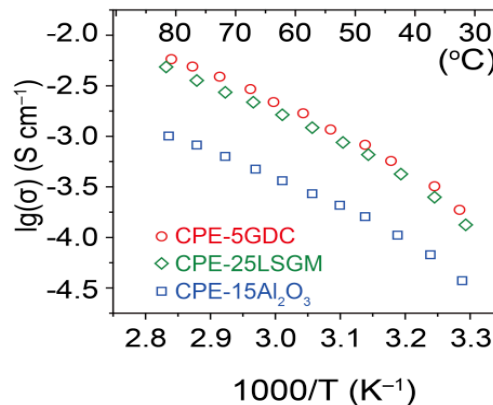
Z. Bao and Y. Cui

# Technical Accomplishments : New Polymer Electrolytes and Protection layers

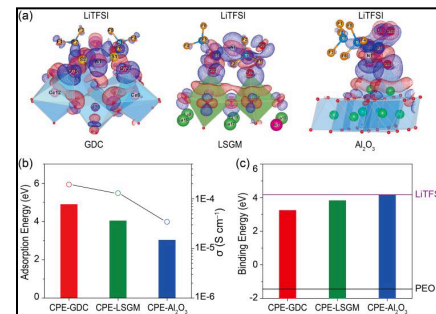
## Polymer/polymer composite ultrathin electrolyte



## Polymer/inorganic composite electrolyte



GDC: Gd:CeO<sub>2</sub>  
LSGM: La<sub>0.80</sub>Sr<sub>0.20</sub>Ga<sub>0.80</sub>Mg<sub>0.20</sub>O<sub>3-X</sub>



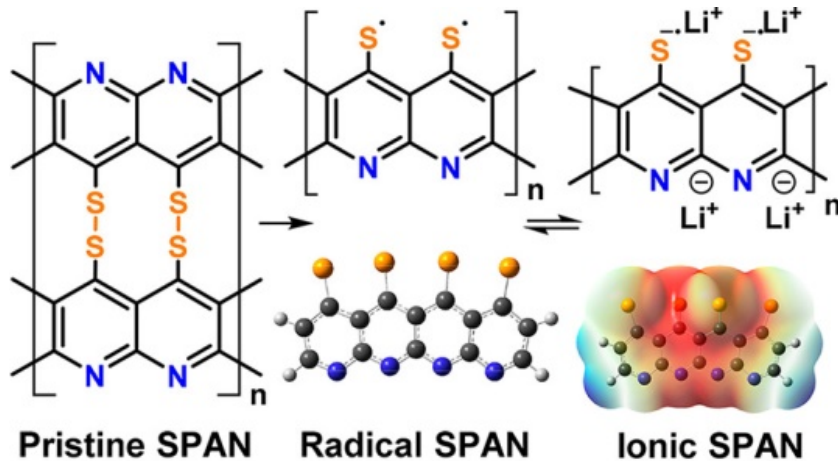
J. B. Goodenough





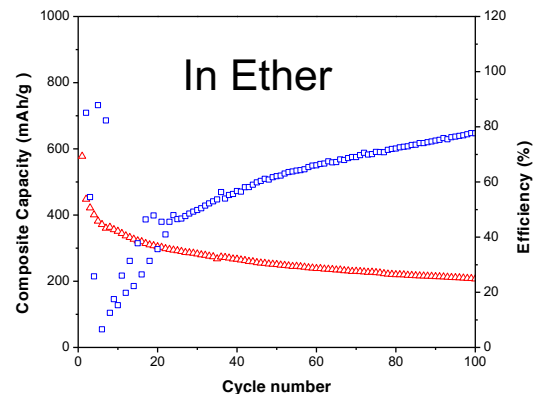
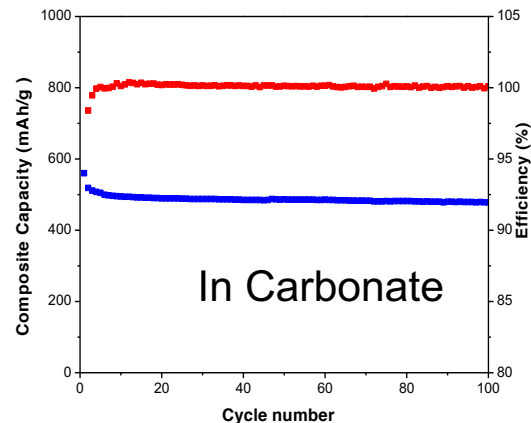
# Technical Accomplishments:

## Stable Li/SPAN Cell to support 250 Wh/kg Pouch Design

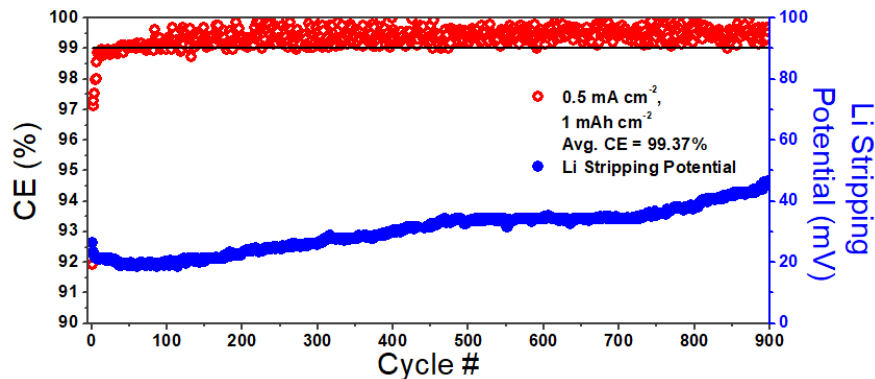


Wang, ACS Energy Lett. 3, 289

- SPAN is a low-cost, highly stable S-based cathode
- SPAN cycles well in carbonate, not ether

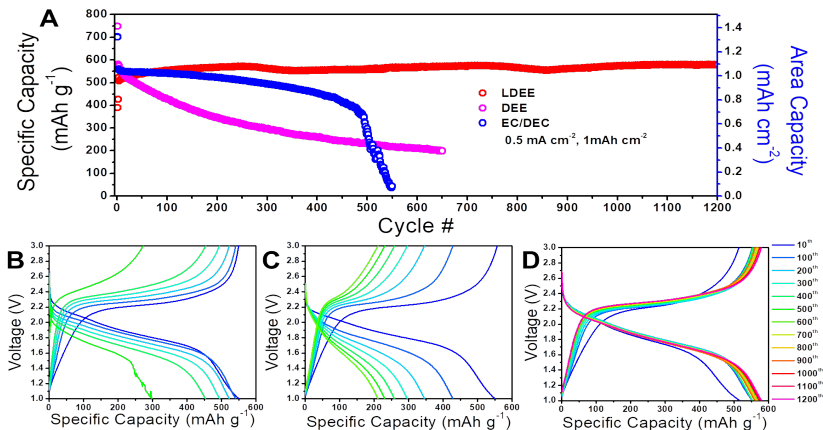
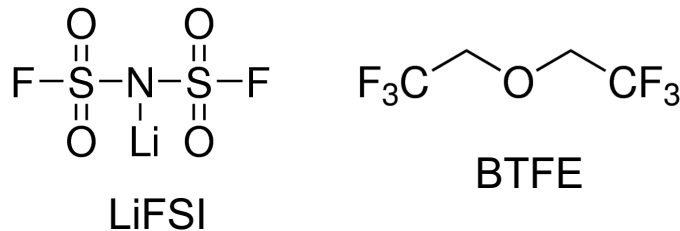


# New Electrolyte That Enables Both Li and SPAN



- Avg. CE of 99.37% for 900 cycles

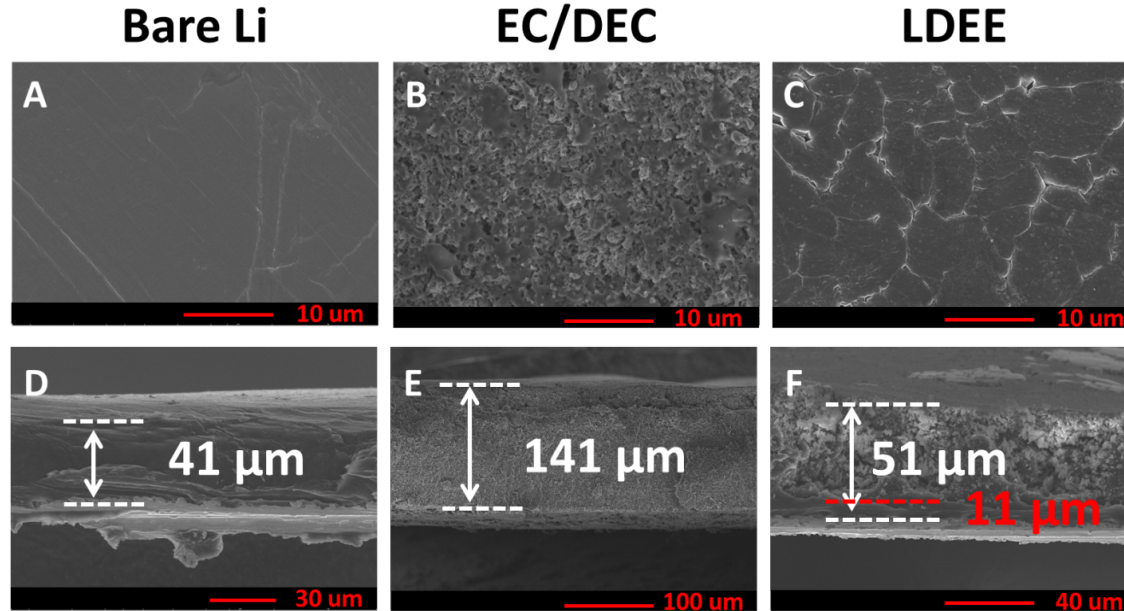
LDEE: 1.8 M LiFSI/DEE-BTFE



- 1200 cycles with no capacity decay

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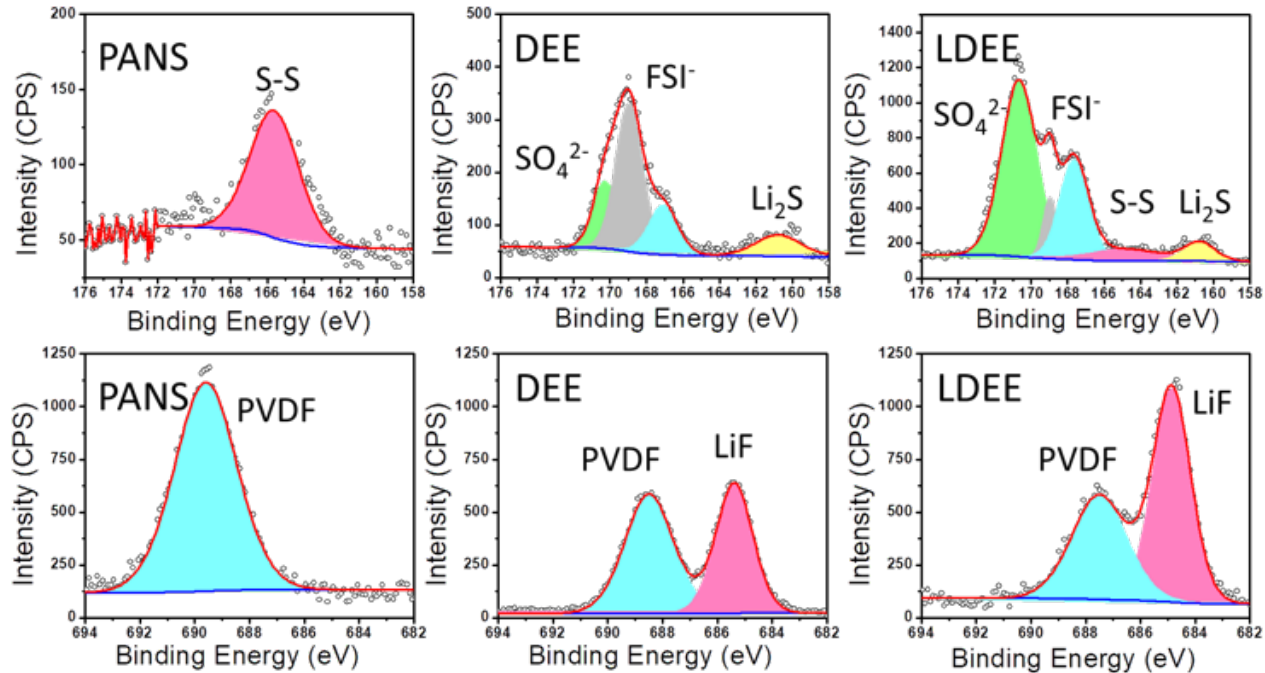
# Lithium Morphology After Cycling



After 62 cycles against SPAN

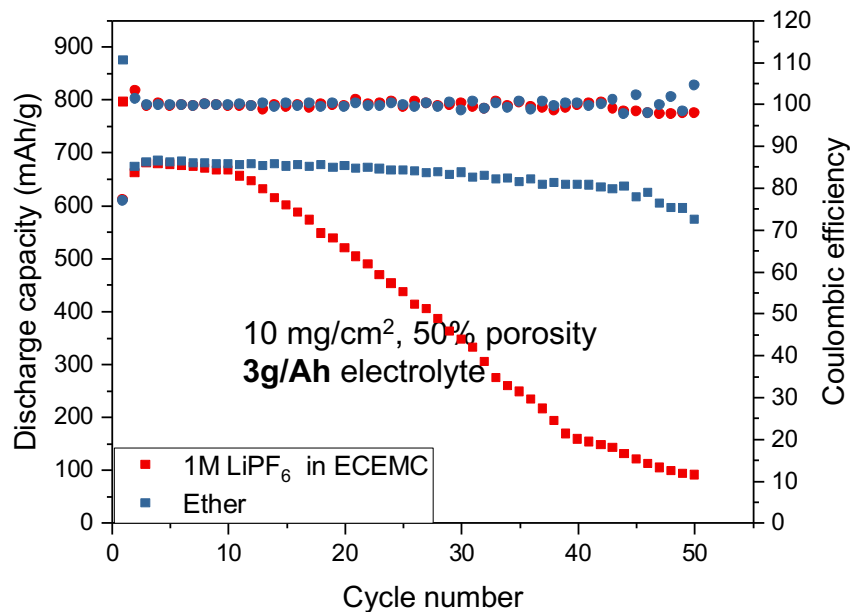
**LDEE enables the best Li morphology**

# Surface Chemistry of Cycled SPAN

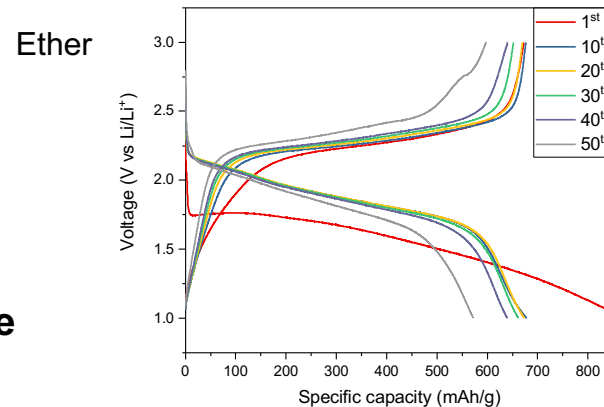
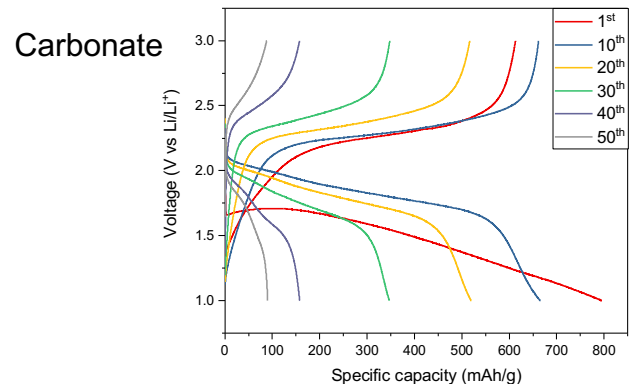


**LDEE promotes the formation of a LiF-rich CEI to protect SPAN**

# Lean Electrolyte With Thick SPAN Cathode



The benefits of using advanced ether electrolytes are more significant under lean electrolyte conditions



# Responses to Previous Year Reviewers' Comments

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- Not reviewed

# UCSD Partners and Collaborators

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- [Brookhaven National Laboratory](#): in-situ XRD and PDF
- [Pacific Northwest National Laboratory](#): Pouch cell design, fabrication, and testing
- [Texas A&M University](#): Computational study of SPAN structures
- [University of Washington](#): Electrochemical modeling of advanced electrode architectures
- [Binghamton University](#): NMC811 data exchange
- [UT-Austin](#): synthesis of NMC811 materials

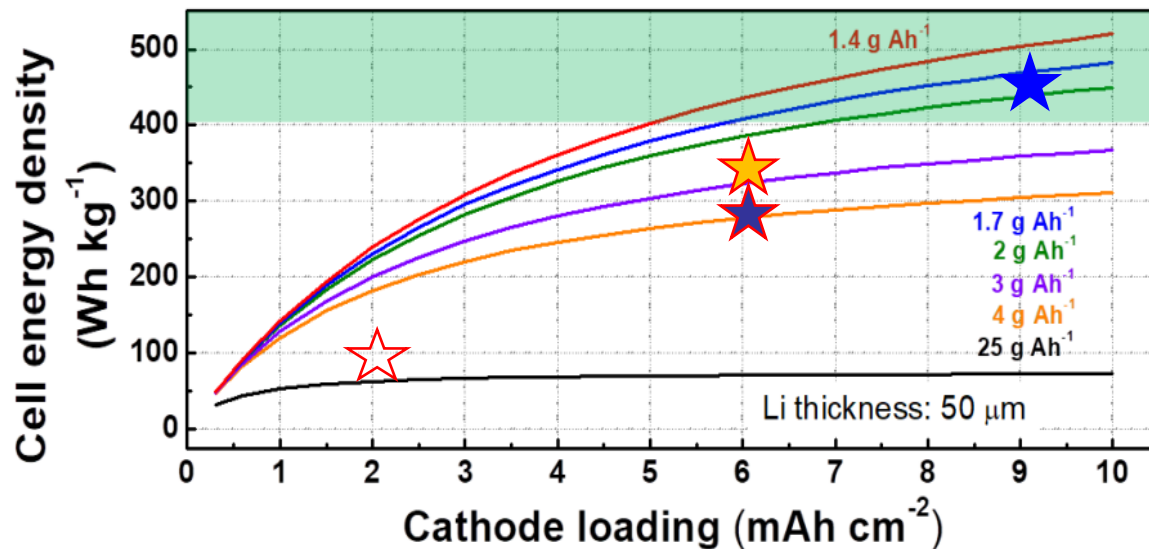
## Remaining challenges for Keystone 2-Li metal

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- **Will 3D Li electrodes make a difference in pouch cells?**
  - How to translate coin cell performance into pouch cells?
  - Are 3D anodes scalable?
- **Can solid electrolyte/protective coating push Li efficiency higher?**
  - Is protective coating synergistic with electrolytes and 3D architectures?
  - How do protective layers work? Do they swell? Do they have selective ion transport? How durable during cycling?



# Remaining Challenges Keystone 2-Li/Thick SPAN



Credit: PNNL  
Based on 800 mAh/g capacity

- Can we further increase loading without losing power?
- Can we raise the SPAN specific capacity?
- Will the results translate into pouch cells?

## Proposed Future Work-Keystone 2

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- Optimize and scale up 3D Li anodes and evaluate them in pouch cells with design parameters (areal specific capacity) that can support Battery500 cell level deliverables;
- Characterize protective coating layers/solid electrolytes after cycling to understand their operating mechanisms and failure modes; Continue to develop mechanically durable, ion conducting materials;
- Determine the maximum sulfur content/capacity of SPAN material; Engineer thick cathodes to support pouch cell design goals; Demonstrate pouch cell performance.

## Keystone 2 Summary

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- Several 3D Li anodes have been fabricated with loading and cycling performance supporting 300-350 Wh/kg cell designs;
- Polymer protective coatings have been developed based on a set of design rules;
- Thin polymer and composite electrolytes have been developed that can serve as lithium metal protection layers
- A Li/SPAN cell enabled by a new electrolyte has shown promising cycle life and potential for high areal loading and lean electrolyte operation